R E M A R K S

The application has been amended so as to place it in condition for allowance at the time of the next Official Action.

Claims 18-40 are present in the application.

Claims 18-23 are withdrawn from further consideration as being drawn to an invention non-elected with traverse.

Claim 24 has been amended.

Claim 27 has been rewritten in independent form.

Claim 27 was objected to under 37 CFR §1.75(c) as being in improper dependent form.

Claim 27 has been rewritten in independent form.

In view of the above amendment to claim 27, Applicants respectfully request reconsideration and withdrawal of the objection to the claim under 37 CFR §1.75(c).

Claims 24-40 were rejected under 35 USC §112, second paragraph, as being indefinite.

Claims 24-40 were also rejected under 35 USC §112, first paragraph, as containing subject matter which was not described in the specification in such a way as to reasonably convey to one skilled in the relevant art that the inventors, at the time the application was filed, had possession of the claimed invention.

Applicants respectfully traverse these rejections.

Normally, in the development of a new material, the material is first made and then characterized and finally used

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based upon its properties. This is a time and labor consuming method. On the other hand, it would be very much preferred if functional material properties could be predicted in a simple way. This is the main object of the present invention, where the nano pore size of elemental carbon can be predicted in certain cases.

Thus, the present invention is directed to the use of a simple calculation to predict nanoporosity of a synthesized elemental carbon material and to produce graphite like pure carbon of predetermined nano pore size in a direct and efficient way. Using this unique nanoporous carbon, with pores formed at an atomic level by three dimensionally connected graphite sheets and created by the decomposition of ordered metal carbide crystal structures, porous articles like electrodes can be made with porosity on two very different levels. Thus, nanoporosity within the carbon structure itself and an additional micro sized porosity between the carbon species to act as "transport porosity" for the electrolyte, for instance, are produced.

Applicants surprisingly discovered that for certain well-crystallized metal carbide materials a simple relation exists, as the metal atoms can be removed from a crystal/particle without having the size and shape of its original geometrical form changed. In other words, metal is removed leaving nanoporosity uniformly distributed throughout the remaining elemental carbon.

This is the formal background why such simple and well known parameters as the number of carbons in the metal carbide molecule unit and atomic weights of the elements involved can be used to calculate the molecular mass of the metal carbide. The atomic/molecular mass of carbon is the same (12). In the formulae also the known density of the crystalline metal carbide and elemental carbon, being 2.2 g/ccm, are included.

Elemental carbon is known to crystallize in two major forms, i.e., diamond and graphite. The densities for crystallized carbon of these forms are very different and well known in physical/chemical Tables. However, diamond is a crystal form that is thermodynamically stable only at extremely high pressures. Thus, in the present invention, the elemental carbon that forms is the normal low-pressure form graphite.

In technical literature the term "carbon" is often misused to describe any pure carbon material, as "carbon" being a technical commodity for many purposes. Thus, such "carbon" might have any density depending on its manufacture and resulting microstructure, pores or faults. However, in the present invention Applicants are using a chemical reaction of well-crystallized metal carbide compounds and the resulting elemental carbon can only have the density of the normal low-pressure graphite form of 2.2 g/ccm, as shown in Example 1 beginning on page 14 of the specification.

Porosity at two such different levels as nano- and

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micro-porosity requires different analytical techniques for determination. The extremely small nano pores require a gas that have atoms or molecules smaller or at most of similar size as the nano pores to be determined. Micro pores can be determined by many different ways. Thus, it is possible to distinguish between the two different pore sizes and volumes even in a produced article as shown in Example 1.

By using the formula/relationship set forth in claim 24, it is possible by calculation to predict the nanoporosity of the produced carbon using the recited crystalline metal carbides.

Claim 28 recites how to produce a desired transport porosity in the final carbon product by the paralysis step recited in claim 24.

Claim 24 has been amended to recite "a porous carbon article having a desired nanoporosity". The metal carbides used are crystallized and contain no nanoporosity.

In the present application crystalline metal carbide particles are employed, as otherwise the relationship of the formulae will not be valid. Using carbide precursors containing a high degree of nanoporosity will influence the results depending on the amount of the precursor porosity and the prediction will be subsequently faulted.

GOLDBERGER 4,543,240 indicates that SiC can have many different porosities, even though it is still SiC. Firstly, it can be noted that GOLDBERGER relates to a technique for

production of metal carbides and the major part regards manufacture of coarser SiC particles. However, column 2 of GOLDBERGER indicates that so called fine precursor particles, less than approximately 10 microns in diameter, can be used to form SiC of "extremely small" particle size. Secondly, in column 7 of GOLDBERGER, it is added that micro-cracks and micro-porosity can make particles of substantially higher surface area.

Forming particles with defects that are micro-sized is not the same as nano-sized porosity. Micron sized particles can still be SiC, despite the presence of micro-cracks or micro-pores. Substantial areas between the defects will be crystalline SiC and follow the relationships mentioned in the application by removing silicon and forming nano pores on a 3 to 4 magnitude lower level than these defects. There is no logical argument why widely differing nanoporosity will be achieved.

It should be stressed that it is not possible to pick an element off the periodic table, as assumed by the Examiner, to obtain any wished nanoporosity. Only the specified elements that form well-defined crystalline carbides can be used. The formulae relates to the collapse of crystal structures where carbon is placed in certain positions, often forming graphite like planes extending through the atomic structure, and that upon collapse form three-dimensionally connected graphite planes resulting in the nanoporosity.

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The number of carbons in the MeC_x molecule, the molecular mass of carbide and carbon, and the specific densities of carbide and carbon (graphite) will give a unique number for R from which X will follow.

Example 1 describes a product containing both nano and micro porosity. The nanoporosity is from the reacted metal carbide to carbon. The micro porosity relates to the transport channels or transport porosity of the article. This is a good example of how to obtain two very different size levels of porosity in the same article.

On page 4 of the Official Action, Applicants were requested to perform certain experiments and to report the results of said experiments.

Attached to the present amendment are seven pages by one of the Applicants, Alla Grechinskaya, explaining why the results regarding Mo contained in the first version of the specification and taken out during the PCT-proceedings were wrong. From this explanation, Applicants submit that it was the experimental measurement that was miscalculated.

In regard to the peak of one micron shown in Figure 2, such peak refers to the transport porosity and not the nanoporosity.

In view of the above amendments and remarks, Applicants respectfully request reconsideration and withdrawal of the

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rejections of claims 24-40 under 35 USC §112, first and second paragraphs, set forth in the Official Action.

Claims 24-40 were rejected under 35 USC §103(a) as being unpatentable over AVARBZ et al. 5,876,787.

Statement Concerning Common Ownership

Application No. 09/424,760 and US patent No. 5,876,787 to AVARBZ et al. were, at the time the invention of Application No. 09/424,760 was made, owned by Frenton Limited.

In view of the above statement, Applicants respectfully submit that as per 35 USC §103(c), AVARBZ et al. is disqualified from being used in a rejection under 35 USC §103(a) based on USC 35 USC §102(e) against the claims of the present application.

In view of the above statement and remarks, Applicants request reconsideration and withdrawal of the rejection of claims 24-40 under 35 USC §103(a) as being unpatentable over AVARBZ et al.

In light of the above remarks, Applicants believe that the present application is in condition for allowance and an early indication of the same is respectfully requested.

If the Examiner has any questions or requires clarification of any of the above points, the Examiner is requested to contact the undersigned Attorney so that this application may continue to be expeditiously advanced.

Attached hereto is a marked-up version of the changes made to the claims. The attached page is captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE."

Respectfully submitted,

YOUNG & THOMPSON

By _____

Benoît Castel
Attorney for Applicants
Registration No. 35,041
745 South 23rd Street
Arlington, VA 22202
Telephone: 703/521-2297

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VERSION WITH MARKINGS TO SHOW CHANGES MADE

Claim 24 has been amended as follows:

--24. (twice amended) A method for producing a porous carbon article comprising the steps of:

selecting powders of at least one carbide of an element selected from the group consisting of Group III, IV, V and VI of Mendeleyv's Periodic System, the at least one carbide having physical and chemical constants to obtain a <u>porous carbon article having a</u> desired nanoporosity <u>by calculating</u> using the relationship:

$$X = Z*(1-R)/R$$

where X = specified size of desired nanopores and $X \leq 10$nm, nm;

$$Z = 0.65-0.75 \text{ nm;}$$

$$R = \nu M_c \rho_k / M_k \rho_c$$

where

M_c - molecular mass of carbon, g/mole;

M_k - molecular mass of the selected carbide, g/mole;

ρ_k - density of the selected carbide, g/ccm;

ρ_c - density of carbon, g/ccm;

ν - number of carbon atoms in carbide molecule;

forming an intermediate body with transport pores having a size larger than 100 nm by shaping the selected powders[,]<u>;</u>

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heat treating the intermediate body in a medium of gaseous hydrocarbon or hydrocarbon mixtures at a temperature exceeding the decomposition temperature for the hydrocarbon or hydrocarbons until the mass of the intermediate body has increased at least 3% thereby producing a work piece in the form of a rigid carbonaceous skeleton; and

thereafter thermochemically treating the work piece in a medium of a gaseous halogen to produce the porous carbon article having nanopores of a size X.--

Claim 27 has been amended as follows:

--27. (amended) [The method according to claim 24,] A method for producing a porous carbon article comprising the steps of:

selecting powders of at least one carbide of an element selected from the group consisting of Group III, IV, V and VI of Mendeleyv's Periodic System, the at least one carbide having physical and chemical constants to obtain a porous carbon article having a desired nanoporosity by calculating using the relationship:

$$X = Z*(1-R)/R$$

where X = specified size of desired nanopores and X<10nm, nm;

$$Z = 0.65-0.75 \text{ nm};$$

$$R = \nu M_c \rho_k / M_k \rho_c$$

where

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M_c – molecular mass of carbon, g/mole;

M_k – molecular mass of the selected carbide, g/mole;

ρ_k – density of the selected carbide, g/ccm;

ρ_c – density of carbon, g/ccm;

ν – number of carbon atoms in carbide molecule;

forming an intermediate body with transport pores having a size larger than 100 nm by shaping the selected powders;

heat treating the intermediate body in a medium of gaseous hydrocarbon or hydrocarbon mixtures at a temperature exceeding the decomposition temperature for the hydrocarbon or hydrocarbons until the mass of the intermediate body has increased at least 3% thereby producing a work piece in the form of a rigid carbonaceous skeleton; and

thereafter thermochemically treating the work piece in a medium of a gaseous halogen to produce the porous carbon article having nanopores of a size X, and

wherein the intermediate body has a porosity determined with the following relationship:

$$\varepsilon_0 = (1 - \nu_{np}/\Sigma K_i \varphi_i) * 100$$

ε_0 porosity of intermediate body vol%;

where

φ_i – volumetric part of i-th carbide in particle mixture;

ν_{np} – predetermined volumetric part of nanopores in final article;

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$$K_i = 1 - \nu M_c \rho_{ki} / M_{ki} \rho_c$$

where

M_c - molecular mass of carbon, g/mole;

M_{ki} - molecular mass of i-th carbide, g/mole;

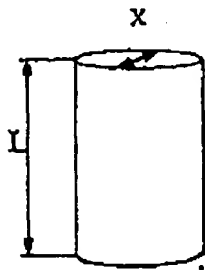ρ_{ki} - density of i-th carbide, g/ccm;

ρ_c - density of cabon, g/ccm;

ν - number of carbon atoms in carbide molecule.--

The size of nanopores is impossible to determine directly by measurements, as they are very small. Even electron microscopy of high resolution cannot be applied in this case. That is why to study nanoporous structure the adsorption methods are used. The adsorption methods of study are based on measurement of adsorption and desorbtion of different substances (for example, nitrogen, benzene) depending on their relative vapor pressure. These methods ensure determination of nanopore volume and surface area of the material. The nanopore volume is estimated by adsorption isotherm at adsorbate relative vapor pressure close to 1. The surface area is calculated using adsorption and desorbtion isotherms and using different mathematical models, for example BET, which is the most acknowledged.

Using ratio of nanopore volume to surface area the pore sizes are determined. The ratio is changed depending on shape of pores. Fig.1 shows the procedure of pore sizes calculation for cylindrical and slit pores. As it can be seen from the Figure the value calculated by cylindrical model has a 2 times difference from the calculated one by slit model.
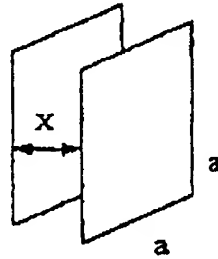
Cylindrical pore   **Slot-like pore**

$$V = \pi X^2 L/4$$
$$S = \pi XL$$

$$S = 2a^2$$
$$V = a^2 x$$

$$\frac{V}{S} = \frac{\pi \cdot x^2 \cdot L}{4 \cdot \pi \cdot x \cdot L} = \frac{x}{L} = \frac{x}{4} \qquad \frac{V}{S} = \frac{a^2 \cdot x}{2 \cdot a^2} = \frac{x}{2}$$

$$x = \frac{4V}{S} \qquad\qquad\qquad x = \frac{2V}{S}$$

Fig. 1. The scheme of calculation of nanopore size for cylindrical and slit-like pore.

In the preliminary calculations of the material pore sizes using the formula (1) we assume that carbon material has slit pores. That is why in the calculations of pore sizes from experimental data it is necessary to use a model of slit pores.

In this connection the more concrete data are specified in the Examples 1-3 as well as additional measurement results.

1. Table 1 shows the values of pore volume and surface area determined by different processing methods of adsorption and desorption isotherms. These data were taken from the KTH reports on measurement and processing of experimental data.

Table 1

Date of measure ment	Type of material	Pore volume V, ccm/g			Surface area S, sqm/g		
		«single point» at $p/p_s = 0.98$	BJH adsorption	BJH desorp-tion	BET	BJH adsor-ption	BJH desor-ption
30.06-3.07.97	TiC #0630	0.42	0.04	0.04	931	16.5	9.13
12-13. 09.97	Mo$_2$C #0912	0.81	0.67	0.84	518	518	958
29-31. 10.97	TiC + Mo$_2$C #1028	0.52	0.18	0.33	892	192	363

As it is known a pore volume determined by a single point of the adsorption isotherm at $p/p_s = 0.98$ represents a total volume of all adsorption pores, while a pore volume determined by BJH method is a volume of pores of size range 1,7 to 300 nm. Surface area calculated by BET method takes account of the surface of all adsorption pores and the surface area calculated by BJH method - only surface of pores of size range 1,7-300 nm.

2. Table 1 shows that «titanium carbide» samples have mainly pores of size less than 1,7 nm (as the pore volume determined by BJH method is very small). While almost all pores in the samples based on Mo$_2$C are of size greater than 1,7

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Table 2

Type of material	$V = V_P - V_{BH}$, cm³/g		$S = S_{BET} - S_{BH}$, m²/g		$X_A = \dfrac{2V_A}{S_A}$, (nm)	$X_D = \dfrac{2V_D}{S_D}$, (nm)	V_{BH}, cm³/g		S_{BH}, m²/g		$X_A = \dfrac{2V_A}{S_A}$, (nm)	$X_D = \dfrac{2V_D}{S_D}$, (nm)
	$V_A = V_{SP} - V_{BH}$ adsorption	$V_D = V_{SP} - V_{BH}$ desorption	$S_A = S_{BET} - S_{BH}$ adsorption	$S_D = S_{BET} - S_{BH}$ desorption			$V_A = V_{BH}$ adsorption	$V_D = V_{BH}$ desorption	$S'_A = S_{BH}$ adsorption	$S' = S_{BH}$ desorption		
TiC	0.38	0.38	914	922	0.83	0.83	-	-	-	-	-	-
Mo₂C	-	-	-	-	-	-	0.67	0.84	518	958	2.58	1.75
TiC + Mo₂C	0.34	0.19	700	529	0.97	0.72	0.18	0.33	192	363	1.88	1.82

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nm (as the volume by BJH method is close (by desorption curve – coincides) to that determined by «single point» method).

Pore size, X, in the nanomaterials using Slit Model of Pores is calculated by the following formula (see fig1)

X = 2V/S

For TiC based samples the values of pore volume and specific surface are used those for pores of size less than 1,7 nm, i.e. $V = V_{SP} - V_{BJH}$ and $S = S_{BET} - S_{BJH}$.

Pore size of carbon of molybdenum carbide is calculated by the formula X=2V/S, using data of BJH method, because all pores of the material are considered to be of size greater than 1,7 nm. The calculated results are presented in Table 2.

3. While calculating pore sizes of carbon produced of the mixture it is necessary to take account of the fact that the material has two types of pores. One group of pores is formed as a result of chlorinating the particles of molybdenum carbide, and the other - of titanium carbide. Pore size in the particles produced of Mo_2C can be calculated by the formula: X=2V/S using BJH data. Size of pores formed after chlorinating the titanium carbide was calculated using the following: pore volume was estimated as a difference of pore volumes measured by «single point» at $p/p_s = 0.98$ and BJH method (see Table2); surface area was calculated as a difference of these values estimated by BET and BJH method. The calculation results are presented in Table 2.

4. The mean pore size of the carbon formed of titanium carbide according to Table2 (by 4 values) is 0.84 nm and of molybdenum carbide (by 4 values) - 2,00 nm.

5. We should pay attention on parameter R of formula (1) of description. This parameter is calculated by using the value of carbon skeleton density ρ_c. Carbon skeleton that is formed as a result of carbides chlorination characterizes by «disordered» graphite-like structure. Its density is different from the density of

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monocrystal graphite and may be rather lower. The experimental results show that the value of density lies in the range of 2.0-2.2 g/ccm.

Thus we have an inaccuracy in the description. Example 1 presents the prior calculation of nanopore size for carbon from titanium carbide with using carbon density value of 2.2 g/ccm. In this case the range of nanopore sizes for carbon from TiC is 0.8-0.92 nm. In table 1 of description presented the results of calculation with carbon density of 2,1 g/ccm which was measured for this samples by helium picnometer. So the range of nanopore sizes for TiC is of 0.74-0.85, and for Mo_2C - 1.95-2.25.

Example 4.

The example of producing an article in a tablet form of d=20mm, h=6 mm with nanopore volume of 0.35 ccm/ccm uniformly distributed throughout the article volume suitable for adsorption of organic substances, out of powder of boron carbide was chosen.

In order to produce the predetermined volume (V_{np} = 0.35 ccm/ccm) before pressing the needed porosity of the work-piece was calculated using expression (3), where M_{B4C} = 55.2 g/mole, ρ_{B4C}=2.51 g/ccm, R=0.253; φ_1=1, n=1, as follows: ε_0=53 %.

The density of carbon for this material was of 2.16 ccm/g.

The amount of boron carbide powder necessary to produce the intermediate body having the predetermined porosity was calculated as in Example 1. In this case the powder article weight was calculated to: m=4.73 g.

The needed mass change of the work-piece during pyrocarbon deposition was calculated by formula (4), assuming a transport porosity of 30 % vol., to Δ m =17.8 %.

Mixture preparation and pyrocarbon deposition was carried as in example 1. The chlorination process was carried out at 950°C.

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The nanopore volume of obtained material was measured by adsorption of benzene. The surface area was measured by BET method. Nanopore size was calculated according to equation: $2V_{sp}/S$. All results are presented in table 3.

As we see from table 3 and 4 all experimental values of nanopore sizes are lies in the interval of calculated values. It proves that all precursor materials that was chosen for nanoporous carbon obtaining are follow the model structure for dependence (1).

Table 3.

Precursor material	Nanopore volume, ccm/g (measured by benzene in CKLM)	Surface area, sqm/g (mesured in KTH: Jie Zeng)	Nanopore size, nm	
			(calculated by formula 1)	(calculated by experimental by data)
B_4C (Bor-B1/98)	0.75	713.8 (material report No 61, 12.02.99)	1.92-2.22	2.10

In addition we present some data about carbon from Mo_2C (sample from the batch # Mo-7B/98IO – a tablet with d= 20 mm and h= 1 mm) and carbon made of SiC - A (a tablet with d≈20 mm and h = 1 mm) that was measured by gas porosity in KTH at October -December of 1996. (For information)

Table 4

Precursor material	Nanopore volume, ccm/g	Surface area, sqm/g	Nanopore size, nm	
			(calculated by formula 1 at carbon density 2.1g/ccm)	(calculated by experimental by data)
NP- Mo_2C	(measured by benzene in CRIM) 0.93	(mesured in KTH: Jia Zeng, fax 99-0.3-29) 972	1.95-2.25	1.91
NP-SiC Type A	measured by gas porosimetry, report No 1		0,77-0,89	
	0.567 96-10-03	1201		0.95
	0,553 96-10-04	1208		0,91
	0,537 96-12-06	1245		0,86
	0,540 96-12-07	1245		0,87

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